

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of
Hiroaki MURASE et al.

Serial No. 10/501,890 Group Art Unit 1621

Filed May 19, 2006

Examiner Mr./Mrs. GALE, KELLETTE

For: METHOD FOR PRODUCING FLUORENE DERIVATIVE

DECLARATION UNDER RULE 1.132

Honorable Commissioner of Patent and Trademarks,
Washington, D.C.

Sir,

I, Hiroaki MURASE, declare:

That I am a citizen of Japan, residing at 12-4,
Taniguchikakinouchi-cho, Ukyo-ku, Kyoto, Japan;

That I was born on March 28, 1966 in Kyoto and
completed the master courseⁱⁿ in Department of Synthetic
Chemistry, Graduate School of Engineering, Kyoto
University, JAPAN in March 1991;

That I have been employed by Osaka Gas Company
Limited, JAPAN since April 1991, and have been on loan
to Osaka Gas Chemical Company Limited, JAPAN since
April, 2007;

That I have been engaged in organic synthesis at

Osaka Gas Company Limited; and

That I currently work at Osaka Gas Chemical Company Limited, Fine Material Production Center, Fine Materials Department, as General Manager; and

That the following experiments were conducted under my direct supervision;

EXPERIMENT

I carried out the following experiments for clearly demonstrating the effects of the present invention (reactant concentration effects).

Incidentally, the following examples and comparative examples were performed in the same manner as Example 1 described in the specification except for varying the amount of hydrochloric acid aqueous solution.

In the following examples and comparative examples, the yield of a fluorene derivative was calculated based on a proportion (molar ratio) of the fluorene derivative relative to fluorenone. Moreover, the "b" value was determined (or calculated) from a transmittance measured at a wavelength of 380 to 780 nm by a visible and ultraviolet absorption apparatus (manufactured by Hitachi, Ltd.).

(Example A)

In a 2 L glass vessel equipped with a stirrer, a

cooler and a thermometer were charged fluorenone (75 g) having a purity of 99% by weight, o-cresol (270 g), β -mercaptopropionic acid (8.5 g), and 36% by weight of hydrochloric acid aqueous solution (47 g). The mixture was subjected to a reaction with stirring under an atmosphere of an inert gas at 25°C for 6 hours, followed by at 35°C for 11 hours. The analysis of the reaction product by HPLC showed that the remaining amount of fluorenone was not more than 0.1% by weight.

After toluene (300 g) and water (80 g) were added to the resultant reaction solution, an aqueous solution containing sodium hydroxide (32% by weight) was added to the mixture for neutralization to approximately pH 7, then the resulting water layer was removed. The organic layer was heated to 80°C, and washed with water (80 g) three times.

After collecting toluene (300 g) by distillation under a reduced pressure, a mixture (500 ml) containing toluene and acetone at a mixing ratio (weight ratio) [toluene/acetone] of 1/4 was added to the organic layer, and stirred at 70°C for one hour. Then, the resultant mixture was cooled to 10°C for crystallization to give an object product, 9,9-bis(4-hydroxy-3-methylphenyl)fluorene (141 g, yield 90%).

The "b" value of thus obtained fluorene derivative was 1.5 (colorless and transparency). The fluorene derivative can be therefore used as a raw

material of polymer without further crystallization operation.

(Example B)

A reaction was carried out in the same manner as Example A except for using 36% by weight of hydrochloric acid aqueous solution (4.7 g) instead of 36% by weight of hydrochloric acid aqueous solution (47 g). The analysis of the reaction product by HPLC showed that the remaining amount of fluorenone was not more than 0.1% by weight.

The same operation as Example A was carried out to the resultant reaction solution, and then 9,9-bis(4-hydroxy-3-methylphenyl)fluorene (135 g, yield 86%) was obtained as an object product.

The "b" value of thus obtained fluorene derivative was 2.9 (colorless and transparency). The fluorene derivative can be therefore used as a raw material of polymer without further crystallization operation.

(Comparative Example A)

A reaction was carried out in the same manner as Example A except for using 36% by weight of hydrochloric acid aqueous solution (118 g) instead of 36% by weight of hydrochloric acid aqueous solution (47 g). The analysis of the reaction product by HPLC showed that the remaining amount of fluorenone was more than

30% by weight because of a large amount of water, which showed an inhibitory action on reaction, present in the reaction system.

The same operation as Example A was carried out to the resultant reaction solution, and then 9,9-bis(4-hydroxy-3-methylphenyl)fluorene (36 g, yield 23%) was obtained as an object product.

The "b" value of thus obtained fluorene derivative was 10.5 (colorless and transparency). The fluorene derivative is not suitable for a raw material of polymer.

(Comparative Example B)

A reaction was carried out in the same manner as Example A except for using 36% by weight of hydrochloric acid aqueous solution (1.18 g) instead of 36% by weight of hydrochloric acid aqueous solution (47 g). The analysis of the reaction product by HPLC showed that the remaining amount of fluorenone was more than 30% by weight because of an insufficient amount of hydrogen chloride for the reaction.

The same operation as Example A was carried out to the resultant reaction solution, and then 9,9-bis(4-hydroxy-3-methylphenyl)fluorene (24 g, yield 15%) was obtained as an object product.

The "b" value of thus obtained fluorene derivative was 6 (colorless and transparency). The

fluorene derivative is not suitable for a raw material of polymer.

In Examples A and B, Comparative Examples A and B, the evaluation results of the yield and the "b" value were described in Table A. Incidentally, the evaluation of Example 1 of the present specification was described for comparison.

Table A

	Example 1	Example A	Example B	Com. Example A	Com. Example B
the mercaptocarboxylic acid/hydrogen chloride (weight ratio)	1/0.87	1/2	1/0.2	1/5	1/0.05
yield (%)	89	90	86	23	15
"b" value	1.3	1.5	2.9	10.5	6.0

EVALUATION

As apparent from the above results, the fluorene derivatives of Examples A and B had high yield and transparency similar to the results of Example 1. Contrarily, the fluorene derivatives of Comparative Examples A and B had low yield and transparency.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Signed this 21st day of August, 2008

Hiroaki Murase

Hiroaki MURASE